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(54) An ion exchange membrane and electrode assembly for an electrochemical cell

(57) This invention provides a solid polymer ion exchange membrane/electrode assembly, or an electrode/solid polymer ion exchange membrane /electrode assembly, for an electrochemical cell, which consists of planar layers of materials intimately joined together to form a unitary structure. The layers are joined together by solid polymer ion exchange resin present in at least one of each pair of adjacent layers, at least one of said layers comprising porous expanded polytetrafluoroethylene. The unitary assembly can be used in an electrochemical cell such as a battery, electrolytic reactor, or fuel cell.

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a release surface, oven-dried to form a thin layer, and, after sufficient layers have been added to form the film, removed and hot pressed to an ion exchange membrane. An alternative method is also disclosed in which a different form of the ion exchange resin is solubilized in the ink mixture, the electrode ink is applied to the surface of an ion exchange membrane, heated and dried to form a layer, and, after sufficient layers have been added to form the film, treat the assembly to convert the ion exchange resin to its use form.

This invention provides an electrode/solid polymer ion exchange membrane assembly for an electrochemical cell comprising planar layers of materials intimately joined together to form a unitary structure. The layers are intimately joined together by a bond, formed across the layer interface, by solid polymer ion exchange resin present in at least one of each pair of adjacent layers. Each embodiment of the invention has one or more layers supported by at least one preformed support matrix formed of porous polytetrafluoroethylene. The preformed support matrix of polytetrafluoroethylene contains the electrode-forming or ion exchange membrane-forming materials of the layer, and provides strength, reinforcement, and handleability, while substantially preventing migration of the materials into adjacent layers.

A planar article or form, as used herein, is an article or form made so as to have length and width dimensions, or radial dimensions, much greater than the thickness dimension. Examples of such articles include a polymeric film or membrane, a sheet of paper, a textile fabric, a ribbon, or a disc, and the like. It is apparent that, once formed, such articles can be used as an essentially flat article, or wound, folded, or twisted into more complex configurations.

By porous as used herein is meant a structure of interconnected pores or voids such that continuous passages and pathways throughout a material are provided.

One embodiment of the invention is a unitary assembly which comprises a planar composite solid polymer ion exchange membrane comprising at least one preformed membrane-support of porous polytetrafluoroethylene which contains, and is made nonporous by, solid polymer ion exchange resin; and a planar electrode in intimate contact with and bonded to a planar surface of the solid polymer ion exchange membrane by the solid polymer ion exchange resin.

Another embodiment of the invention is a unitary assembly which comprises a planar composite solid polymer ion exchange membrane comprising at least one preformed membrane-support of porous polytetrafluoroethylene which contains, and is made nonporous by, solid polymer ion exchange resin; and two planar electrodes, each electrode in intimate contact with and bonded to a planar surface of the solid polymer ion exchange membrane by the solid polymer ion exchange resin.

Yet another embodiment of the invention is a unitary assembly which comprises a planar composite solid polymer ion exchange membrane comprising at least one preformed membrane-support of porous polytetrafluoroethylene which contains, and is made nonporous by, solid polymer ion exchange resin; and a planar electrode comprising a preformed electrode-support of porous polytetrafluoroethylene containing both a solid polymer ion exchange resin and a catalyst material, and which is in intimate contact with and bonded to a planar surface of the solid polymer ion exchange membrane by solid polymer ion exchange resin.

A further embodiment of the invention is a unitary assembly which comprises a planar composite solid polymer ion exchange membrane comprising at least one preformed membrane-support of porous polytetrafluoroethylene which contains, and is made nonporous by, solid polymer ion exchange resin; and two planar electrodes each of which comprises a preformed electrode-support of porous polytetrafluoroethylene containing both a solid polymer ion exchange resin and a catalyst material and which are in intimate contact with and bonded to a planar surface of the solid polymer ion exchange membrane by solid polymer ion exchange resin.

Other embodiments of the invention are unitary assemblies which comprise a planar solid polymer ion exchange membrane and one or two planar electrodes; each electrode comprising a preformed electrode-support of porous polytetrafluoroethylene containing both a solid polymer ion exchange resin and a catalyst material, and each electrode in intimate contact with and bonded to a planar surface of the solid polymer ion exchange membrane by solid polymer ion exchange resin.

This invention provides a unitary solid polymer ion exchange membrane/electrode assembly. The solid polymer ion exchange membrane comprises at least one preformed membrane-support of porous expanded polytetrafluoroethylene which is filled and made nonporous with solid polymer ion exchange resin. The porous PTFE membrane-support is typically filled by impregnation of a liquid composition containing the ion exchange resin into the membrane-support. The electrode comprises electrode constituents, which include both a solid polymer ion exchange resin and a catalyst material, and a support matrix of polytetrafluoroethylene. The electrode is intimately joined and adhered to the solid polymer ion exchange membrane by a bond formed between the ion exchange resin present in both layers.

As noted above, in the unitary solid polymer ion exchange membrane/electrode assembly of the invention, the composite ion exchange membrane comprises at least one membrane-support consisting of a preformed porous film of polytetrafluoroethylene. Electrodes of the assembly preferably also comprise an electrode-support consisting of a preformed porous film of polytetrafluoroethylene. However, other electrode structures, also inclusive of a polytetrafluoroethylene support matrix, preferably an expanded polytetrafluoroethylene support matrix, can be obtained by mixing together elec-

used. Surfactants may also be included in the liquids to facilitate mixing and dispersion of the materials forming the liquid compositions.

As with the ion exchange resins, no particular limitations are placed on the catalyst materials so long as they are amenable to impregnation into and retention by the membrane-support and electrode-support films. Any particulate material, or powder, acting as a catalyst can be used, and will be selected according to the application intended. Examples include, but are not limited to, lead dioxide for ozone generating electrodes, platinum or platinum alloys for hydrolytic electrodes, platinum or platinum alloys supported on carbon black, and the like.

The catalyst materials and a solid polymer ion exchange resin are combined in a liquid mixture for impregnation into an electrode-support film, or for surface coating or impregnation into an electrode structure. This can be done, for example, by dispersing the catalyst powder in solvents such as those described above, and then adding ion exchange resin, or a liquid composition containing ion exchange resin, to form the liquid mixture. If desired, it is also possible to include a fluoropolymer, such as PTFE, tetrafluoroethylene/(perfluoroalkyl) vinyl ether copolymer (PFA), or tetrafluoroethylene/hexafluoropropylene copolymer (FEP), in such liquid mixtures to enhance water repellency in the electrode structure. It is also possible to include a pore-forming agent, such as ammonium bicarbonate, sodium chloride, or calcium carbonate, which is removed after formation of the membrane, for example, by heating or leaching, to create voids to improve gas diffusivity.

Catalyst materials can also be introduced into an electrode structure as a catalyst precursor. In such a case the liquid mixture to be impregnated into an electrode-support film is a mixture obtained by combining a liquid dispersion of noncatalytic electrically-conductive particles and a liquid composition containing the catalyst precursor and a solid polymer ion exchange resin. That is, it may be a liquid mixture of noncatalytic electrically-conductive particles, a solid polymer ion exchange resin, and a solid polymer ion exchange resin which has a catalyst metal precursor bonded to its exchange groups. For example, carbon black is used as the electrically-conductive particles; the carbon black is dispersed in a liquid composition containing solid polymer ion exchange resin to allow the resin to adsorb onto the carbon black. Catalyst metal anions, such as in a platinum-ammine complex solution, are then added to bring about ion exchange, after which more solid polymer ion exchange resin is added. The ingredients can be mixed simultaneously or added sequentially. When such a mixture is used, the catalyst precursor must be converted to a catalyst by some type of reducing treatment after the solid polymer ion exchange membrane/electrode has been formed. Such reducing treatments include heating and hydrogen reduction, chemical reduction using sodium borohydride, and other reducing treatments known in the art. A highly active catalyst can be obtained with the use of such methods.

In the preparation of a unitary ion exchange membrane/electrode assembly of the present invention, the pores of the porous expanded polytetrafluoroethylene membrane-support film are impregnated with a solid polymer ion exchange resin to obtain a composite membrane that is thin yet has high strength. Impregnation can be accomplished using equipment and methods known in the art, and no particular restrictions are imposed. For example, the porous expanded polytetrafluoroethylene membrane-support film can be dipped or immersed in a liquid composition containing the resin; or the liquid composition may be applied to the surface by brushing or spraying, by screen printing, by roll coating, and the like, after which the solvent is removed. Such methods may be repeated a number of times until the pores are essentially completely filled with the solid polymer ion exchange resin and a nonporous composite film is produced. The solvent can be removed by any convenient method such as air drying, heating in an oven or over heated rolls, and the like. If heating is used, temperatures which can lead to decomposition of the ion exchange resin should be avoided. Due to the strength and handleability of the porous expanded polytetrafluoroethylene film, and its ability to retain the liquid composition containing the ion exchange resin in its porous structure, the composite solid polymer ion exchange resin-filled membrane-support film can be formed separately, and subsequently intimately joined to an electrode structure: or it can also be formed in place on the surface of an electrode or other substrate, for example, by first superposing the porous expanded polytetrafluoroethylene membrane-support film on an electrode structure and then impregnating the membrane-support film, which simultaneously joins it to the electrode.

A preferred structure for the unitary solid polymer ion exchange membrane/electrode assembly of the invention comprises an electrode structure having a preformed electrode-support also consisting of a preformed porous film of polytetrafluoroethylene. The porous expanded polytetrafluoroethylene electrode-support films are impregnated with the liquid mixtures containing catalyst materials and solid polymer ion exchange resin by the same means described above. As with the ion exchange membrane-support described above, the electrode-support film can be impregnated separately; or while on a substrate providing a release surface, or on the surface of a substrate to which it is simultaneously intimately joined, such as, for example, the surface of a collector, a gas diffusion material, an ion exchange membrane, or preferably, a composite solid polymer ion exchange resin-filled membrane-support film.

In the course of impregnation and desolvation/solidification of the liquid mixtures, the solid polymer ion exchange resin causes the catalyst particles to adhere to each other and serves as a binder in fixing the catalyst

about 3 micrometers thick (Gore-Tex® expanded PTFE film, manufactured by Japan Gore-Tex, Inc.), and having a nominal pore size of about 1 micrometer and pore volume of 93 percent was fixed to the surface of the coated electrode sheet. The porous PTFE film was coated and impregnated with the same solution applied to the electrode sheet so as to essentially completely fill the pores of the PTFE film and contact the solution coated on the electrode sheet, after which the composite article was subjected to UV radiation to effect crosslinking, and a unitary solid polymer ion exchange membrane/electrode assembly of the invention was produced.

Example 2

A gas diffusion electrode for a fuel cell was prepared as follows:

An aqueous dispersion of carbon black particles ("Denka Black", supplied by Denka Co.) and PTFE resin particles having a solids concentration 65 wt.% carbon black and 35 wt.% PTFE was prepared. The PTFE was coagulated, and the coagulum of mixed carbon black and PTFE dried. Naphtha was added and mixed into the dried coagulum as a lubricant The lubricated coagulum was ram-extruded to form a tape 2.5 mm thick. The extruded tape was calendered and the thickness reduced to 250 micrometers. The calendered tape was uniaxially stretched (in the longitudinal direction) at a temperature of about 250°C to 5 times its original length, and then again calendered to reduce its thickness by a factor of 5. The porous electrically-conductive gas permeable electrode sheet thus produced was about 50 micrometers thick, had a nominal pore size of about 1 micrometer, and a pore volume of about 78 %.

A collector sheet consisting of 130 micrometer thick carbon paper, supplied from Toray Co., was impregnated with an aqueous dispersion of PTFE. The PTFE-treated collector sheet and gas diffusion electrode sheet were laminated together by application of heat (120°C) and pressure (20 kg/cm^2), after which the laminated assembly was heat treated at 360°C for 10 minutes.

A liquid mixture containing catalyst material and solid polymer ion exchange resin was prepared. The catalyst material was platinum-coated (25 wt.%) carbon black (tradename - Vulcan® XC72), and the solid polymer ion exchange resin was Nafion® perfluorosulfonic acid resin (manufactured by DuPont Co.). A dispersion of 5 grams of Pt-coated carbon black in 40 grams of 2-methyl,1-propyl alcohol was prepared. A liquid composition of isopropyl alcohol containing 9 wt.% Nafion® perfluorosulfonic acid resin was added to the dispersion to provide a liquid mixture having a relative concentration of 30 wt.% perfluorosulfonic acid resin and 70 wt.% Pt-coated carbon. The liquid mixture was applied by brush to the surface of the gas diffusion electrode sheet, thereby forming a solid polymer ion exchange resin/catalyst containing region, and the solvent removed, thus completing the electrode structure.

A porous expanded polytetrafluoroethylene film was fixed on the solid polymer ion exchange resin/catalyst coated surface of the electrode. The PTFE film was 20 micrometers thick, had a nominal pore size of 0.2 micrometer, and a pore volume of 89 %. The porous PTFE film was coated with a liquid composition of isopropyl alcohol containing 5 wt.% Nafion® perfluorosulfonic acid resin (manufactured by DuPont Co.), and air dried. The coating and air drying steps were repeated 5 times until the pores of the PTFE film were essentially completely filled, the layers joined by the solid polymer ion exchange resin present in each layer, the composite membrane-support film became semitransparent, and the surface of the film coated with the solid polymer ion exchange resin. The composite assembly was heat treated at 130°C for 24 hours, and a unitary solid polymer ion exchange resin/electrode assembly was obtained.

A second unitary solid polymer ion exchange resin/electrode assembly was obtained exactly as described above. The membrane-supported ion exchange resin surface of one of the assemblies was coated with a liquid composition of isopropyl alcohol containing 2 wt.% Nafion® perfluorosulfonic acid resin, placed on the membrane-supported ion exchange resin surface of the second assembly and lightly pressed to remove entrapped air, after which the solvent was removed by air drying and the joined assemblies heat treated at 130°C for 24 hours.

The unitary assemblies thus joined formed a larger unitary embodiment of the invention to which further components were joined. The complete assembly described above was mounted and operated as a gaseous fuel cell. Humidified hydrogen was fed on one side of the mounted assembly, and oxygen was fed on the other side at an operating temperature of 80°C. The cell developed a voltage of 0.78 volts at a current level of 1 A/cm^2.

Comparative Example 1

An electrochemical cell assembly was prepared as described in Example 2, except that no membrane-support films were used and the liquid composition of isopropyl alcohol containing Nafion® perfluorosulfonic acid resin was applied directly to the gas permeable electrode sheet. Numerous cracks formed, and partial separation from the substrate occurred.

The electrochemical cell assembly was tested in a fuel cell as described in Example 2, and developed a voltage of 0.67 volts at a current level of 1 A/cm^2.

Example 3

A composite membrane-supported solid polymer ion exchange resin-filled film was prepared separately. The PTFE film was 15 micrometers thick, had a nominal pore size of 0.2 micrometer, and a pore volume of 89 %.

A solid polymer ion exchange membrane (Nafion® 117 perfluorosulfonic acid membrane, manufactured by Dupont. Co.) was sandwiched between the first electrode structure and the second electrode structure, and laminated by application of heat (140°C) and pressure (25 kg/cm^2) to form a unitary assembly of the invention.

A platinum-plated titanium mesh was applied to the surface of the first electrode as a collector, and the unitary assembly and collector sandwiched between ribbed, platinum-plated, stainless steel plates to form an electrochemical cell. Purified water was fed to the ribbed portions, and the cell was operated as an ozone generator by water electrolysis using a solid polymer electrolyte.

Example 8

A dispersion of 5 grams of carbon black/platinum (25 wt.%) particles (from NE Chemcat Co.) in 40 grams of 2-methyl, 1-propyl alcohol was prepared. To the dispersion was added a liquid composition of isopropyl alcohol containing 9 wt.% Nafion® perfluorosulfonic acid resin (manufactured by DuPont Co.) and thoroughly mixed, with the aid of ultrasonic agitation, to form a liquid mixture, having a relative concentration of 25 wt.% ion exchange resin and 75 wt.% carbon black supported platinum.

A collector sheet consisting of 230 micrometer thick carbon paper, manufactured by Toray Co., was impregnated with an aqueous dispersion of PTFE to develop water repellency, and then heat treated at 360°C for 10 minutes. A porous expanded polytetrafluoroethylene electrode-support film (thickness - 16 micrometers; pore volume - 94%; IBP - 0.12 kg/cm^2) was fixed to the surface of the carbon paper. The liquid mixture was applied by brush to impregnate the pores of the electrode-support film, after which the solvent was removed by air drying. The composite structure was heat treated at 120°C for 24 hours, thus completing a first electrode.

A porous expanded polytetrafluoroethylene membrane-support film (thickness - 20 micrometers; pore volume - 93%; IBP - 0,15 kg/cm^2) was fixed to the ion exchange resin/catalyst impregnated surface of the first electrode. The porous PTFE film was coated by brush with a liquid composition of isopropyl alcohol containing 5 wt.% Nafion® perfluorosulfonic acid resin (manufactured by DuPont Co.), and air dried. The coating and air drying steps were repeated 3 times until the pores of the PTFE film were essentially completely filled and the layers joined by the solid polymer ion exchange resin present in each layer, thus forming a first unitary assembly of the invention.

An aqueous dispersion of carbon black particles ("Denka Black", supplied by Denka Co.) and PTFE resin particles having a solids concentration 60 wt.% carbon black and 40 wt.% PTFE was prepared. The PTFE was coagulated, and the coagulum of mixed carbon black and PTFE dried. Naphtha was added and mixed into the dried coagulum as a lubricant. The lubricated coagulum was ram-extruded to form a tape 2.5 mm thick. The extruded tape was calendered and the thickness reduced to about 300 micrometers. The calendered tape was uniaxially stretched (in the longitudinal direction) at a temperature of about 250°C to 5 times its original length, and then again calendered to reduce its thickness by a factor of 5. The electrically-conductive gas permeable electrode sheet thus produced was about 60 micrometers thick, had a nominal pore size of about 1 micrometer, and a bulk density of 0.51 g/cc. A collector sheet, identical to the collector sheet bonded to the first electrode, was fixed to one surface of the gas permeable electrode sheet.

A porous expanded polytetrafluoroethylene electrode-support film, identical to the electrode-support film of the first electrode, was fixed to the other surface of the gas permemable electrode sheet, and impregnated with the liquid mixture of ion exchange resin/catalyst particles, and heat treated as described above, thus forming a second electrode.

A porous expanded polytetrafluoroethylene membrane-support film, identical to the membrane-support joined to the first electrode, was fixed to the ion exchange resin/catalyst impregnated surface of the second electrode, and was impregnated with the same materials and in the same manner, thus forming a second unitary assembly of the invention.

A small amount of the same liquid composition of isopropyl alcohol containing 5 wt.% Nafion® perfluorosulfonic acid resin described above was then applied by brush to the surface of the Nafion® resin-filled membrane-support film of the second assembly and the ion exchange resin-containing surfaces of the first and second assemblies were brought together and lightly pressed to remove entrapped air and intimately join the assemblies, after which the solvent was removed by air drying and another embodiment of the unitary assembly of the invention completed.

This embodiment of the invention was mounted and operated as a gaseous fuel cell. Humidified hydrogen was fed on one side of the mounted assembly, and oxygen was fed on the other side at an operating temperature of 80°C. The cell developed a voltage of 0.71 volts at a current level of 1 A/cm^2.

Example 9

A porous expanded polytetrafluoroethylene membrane-support film (thickness - 10 micrometers; pore volume - 83%; IBP - 1.75 kg/cm^2) was superposed on a polypropylene release sheet. The membrane-support film was coated by brush with a liquid composition of isopropyl alcohol containing 5 wt.% Nafion® perfluorosulfonic acid resin (manufactured by DuPont Co.), and air dried at 70°C. The coating and air drying steps were repeated 4 times until the pores of the PTFE film were essentially completely filled, resulting in a virtually trans-

a composite solid polymer ion exchange membrane having first and second planar surfaces: said composite ion exchange membrane comprising at least one preformed membrane-support of porous expanded polytetrafluoroethylene, said membrane-support containing, and made nonporous by, solid polymer ion exchange resin; and
a first electrode having two planar surfaces, one surface of said first electrode in intimate contact with said first surface of said solid polymer ion exchange membrane and bonded to said membrane by said solid polymer ion exchange resin.

2. The assembly as recited in Claim 1 further comprising

a second electrode having a surface in intimate contact with said second surface of said composite solid polymer ion exchange membrane and bonded to said membrane by said solid polymer ion exchange resin.

3. The assembly as recited in Claim 1 wherein said first electrode comprises a preformed electrode-support of porous expanded polytetrafluoroethylene, said electrode-support containing both a solid polymer ion exchange resin and a catalyst material.

4. The assembly as recited in Claim 3 wherein said preformed electrode-support further contains a noncatalytic electrically-conductive material.

5. The assembly as recited in Claim 3 wherein said second surface of said first electrode is in intimate contact with an electrically-conductive gas diffusion material, said first electrode bonded to said electrically-conductive material by said solid polymer ion exchange resin.

6. The assembly as recited in Claim 4 wherein said second surface of said first electrode is in intimate contact with an electrically-conductive gas diffusion material, said first electrode bonded to said electrically-conductive material by said solid polymer ion exchange resin.

7. The assembly as recited in Claim 2 wherein said second electrode comprises a preformed electrode-support of porous expanded polytetrafluoroethylene, said electrode-support containing both a solid polymer ion exchange resin and a catalyst material.

8. The assembly as recited in Claim 7 wherein said preformed electrode-support further contains a noncatalytic electrically-conductive material.

9. The assembly as recited in Claim 7 wherein said second surface of said second electrode is in intimate contact with an electrically-conductive gas diffusion material, said second electrode bonded to said electrically-conductive material by said solid polymer electrolyte resin.

10. The assembly as recited in Claim 8 wherein said second surface of said second electrode is in intimate contact with an electrically-conductive gas diffusion material, said second electrode bonded to said electrically-conductive material by said solid polymer electrolyte resin.

11. A unitary assembly for an electrochemical cell comprising

a solid polymer ion exchange membrane having first and second planar surfaces; and
a first electrode having two planar surfaces;
said first electrode comprising a preformed electrode-support of porous expanded polytetrafluoroethylene, said electrode-support containing both a solid polymer ion exchange resin and a catalyst material;
wherein one surface of said first electrode is in intimate contact with said first surface of said solid polymer ion exchange membrane and bonded to said membrane by said solid polymer ion exchange resin.

12. The assembly as recited in Claim 11 further comprising

a second electrode comprising a preformed electrode-support of porous expanded polytetrafluoroethylene, said electrode-support containing both a solid polymer ion exchange resin and a catalyst material;
wherein one surface of said second electrode is in intimate contact with said second surface of said solid polymer ion exchange membrane and bonded to said membrane by said solid polymer ion exchange resin.

13. The assembly as recited in Claim 11 wherein said preformed electrode-support further contains a noncatalytic electrically-conductive material.

14. The assembly as recited in Claim 12 wherein said preformed electrode-support further contains a noncatalytic electrically-conductive material.

15. The assembly as recited in Claim 11 wherein said second surface of said first electrode is in intimate contact with an electrically-conductive gas diffusion material, said first electrode bonded to said electrically-conductive material by said solid polymer ion

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EUROPEAN SEARCH REPORT

Application Number
EP 95 30 8882

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
X	WO-A-89 06055 (HUGHES AIRCRAFT CO) 29 June 1989 * page 8, line 34; claims 1,9,10,23-25 * * page 11, line 7 - line 32 * * page 18, line 1 - line 32 * * page 23, line 1 - line 15 * * page 25, line 11 - line 19 *	1-4,7,8, 11-14	H01M8/10 C25B9/00
Y	* line 20 - line 34 * ---	5,6,15, 16	
Y	US-A-4 804 592 (VANDERBORGH NICHOLAS E ET AL) 14 February 1989 * column 8, line 62 - column 9, line 54; claims 30-35 * * column 11, line 8 - line 19 * ---	5,6,15, 16	
A	EP-A-0 572 810 (HUGHES AIRCRAFT CO) 8 December 1993 * claims 1,16 * ---	1	
			TECHNICAL FIELDS SEARCHED (Int.Cl.6)
A	AICHE JOURNAL, vol. 38, no. 1, January 1992 NEW YORK US, pages 93-100, XP 000318978 MARK W. VERBRUGGE ET AL 'Composite Membranes for Fuel-Cell Applications' * abstract * * page 93, right column, last paragraph - page 94, left column, paragraph 1 * ---	1	H01M C25B
A	FR-A-2 295 982 (HOOKER CHEMICALS PLASTICS CORP) 23 July 1976 * page 5; claims 1,23 * --- -/--	1	

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	1 April 1996	D'hondt, J

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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